

Thermodynamic Parameters Modeling of Viscous Flow Activation in Ethylene Glycol-Water Fluid Systems

Snoussi, Lotfi

University of Carthage, Thermal Processes Laboratory (LPT), Research and Technology Center of Energy (CRTE_n), Borj Cedria, B.P N°95 - 2050 Hammam-Lif, TUNISIA

Shaik, Babu

Department of Physics, Koneru Lakshmaiah Education Foundation, Vaddeswaram, AP, INDIA

Herráez, Jose V.

Department of Thermodynamics, Faculty of Pharmacy, University of Valencia, Burjassot (Valencia) 46100, SPAIN

Akhtar, Shamim

Department of Chemistry, University of Chittagong, Chittagong-4331, BANGLADESH

Al-Arfaj, Ahlam A.; Ouerfelli, Noureddine*⁺

Department of Chemistry, College of Science, Imam Abdulrahman Bin Faisal University, P.O. Box 1982, Dammam 31441, SAUDI ARABIA

ABSTRACT: Calculation of excess quantities of ethylene glycol + water binary fluid systems at seven absolute temperatures (293.15 to 353.15) K from experimentally determined values of density and shear viscosity were presented in previous work. The examination of related functions beside the quality of correlation from several equations on these experimental values has also been reported. Considering the quasi-equality between Arrhenius activation energy of viscosity E_a and the enthalpy of activation for viscous flow ΔH^* , over with their individual's contribution separately we can define partial molar activation energy E_{a1} and E_{a2} for ethylene glycol with water respectively. Correlation between Arrhenius parameters also shows desirable Arrhenius temperature. Comparison to the vaporization temperature in the fluid vapor equilibrium, and the limiting corresponding partial quantities permit us to predict value of the boiling points of the pure constituents. New empirical equations to estimate the boiling point are developed.

KEYWORDS: Binary fluid systems; Shear viscosity; Arrhenius activation energy; Ethylene glycol; boiling temperature.

*To whom correspondence should be addressed.

+E-mail:nouerfelli@iau.edu.sa

1021-9986/20/3/287-301

15/\$/6.05

INTRODUCTION

Recently we have initiated research work on application of thermodynamic/transport properties of some fluid systems in lately suggested correlation equations [1–13]. After thorough survey of literature and experimental values reported already [14], we have extended our idea of investigating the viscosity Arrhenius behavior along with a new parameter named as Arrhenius temperature (T_A), in ethylene glycol (EG) + water (W) systems from 293.15 K to 353.15 K. Ethylene glycol is an important raw material for the polyester fibers industry [14]. In addition, EG (1,2-ethanediol) is the simplest diol where glycols find prevalent application in the explosives, petroleum, cosmetic, automotive, pharmaceutical, aviation and in some industries, domains, and applications [14]. Generally, data of ethylene glycol (EG) + water (W) binary systems are required for the conception of the heat-exchanger materials used with antifreeze and its production such as in solar energy units, motor vehicles, heat pumps, solar energy, and water heating/cooling systems [14-25].

Furthermore, we considered that the activation energy is practically a thermodynamic state function and partial activation energies have been calculated to discuss the individual contributions of pure fluids in all the component systems under study. The new introduced parameter designated as current Arrhenius temperature (T_{Ai}) related to each pure fluid constituent (i) will lead us to reliably simulate the boiling point value of the pure fluid components in the present system. We can add that the present work indicates various future directions that researchers can undertake to investigate the effect of variable shear viscosity on *heat* and *mass transfer* enhancements of nano fluids suspended in a binary base fluid. In addition, the absolute temperature dependence study of nano fluids and their thermo-physical quantities are very important because it influences the flow characteristics and, through that, influences the heat transfer performance in the cooling system [22-36]. In this way in future works, we will introduce the effects of nano fluids and their characteristics through some nano liquid features, that is, taking into consideration the reports lighting the synthesis, morphological, textural characterization and, thermal stability of nano particles [22-36]. In addition, a comprehensive understanding of the dominant factors and the trends affecting the transport

process and heat transfer in nanofluids is essential for development and design. In this context, new correlation will be developed for the nanofluids attributes in terms of particles volumetric concentration and absolute temperature measurements.

VISCOSITY ARRHENIUS BEHAVIOR

Viscosity Results

In the present study, the shear viscosity (η) and density (ρ) values for EG (1) + W (2) fluid systems with mole composition of EG are used which have been reported in earlier work [14] where these measured quantities values of EG + W binary fluid system were used to calculate the excess molar volumes (V^E) and shear viscosity deviations ($\Delta\eta$) of this binary fluid system, and then fitted to the popular Redlich-Kister (RK) expression. In addition and in the same context, the deviations of shear viscosity $\Delta\eta$ with the mole composition (x_1) of EG and the variation of excess molar volume (V^E) were likewise fitted to the Herráez, Belda, reduced RK, relative reduced (RK) and Grunberg-Nissan equations, and these quantities were interpreted in terms of specific or nonspecific interactions, and the subsequent discussions were evenly reported in previous papers [14,37-40].

The negative values of V^E may possibly be explained as a result of a high number of carbon atoms in EG, leading to low polarity and weakening of the association between EG molecules and hence forming induced dipoles-dipole complexes between EG and W [14,21,22]. Likewise, the negative values of shear viscosity deviation, show that there is an absence of specific interaction between EG and W. Also, it can show absence or disrupt or breaking of hydrogen bond in the system.

In like manner, from the investigation of the Desnoyers *et al.* [41] approach for the treatment of excess thermodynamic quantities for binary fluid systems, we can show that a global outlook of the provenance of the non-ideality in the systems were given by the excess quantity (V^E and $\Delta\eta$) but still can be quite misleading, notably for systems that show strong interactions at very high dilution. According to the original statements of Redlich-Kister [42], Desnoyers *et al.* [41] suggested that the use the reduced RK excess quantity is preferred i.e., the ratio $\Delta\eta/x_1(1-x_1)$ or $V^E/x_1(1-x_1)$ for this purpose. Though EG + W systems are nonelectrolyte solutions, an expansion equation was also adopted and used for the shear viscosity

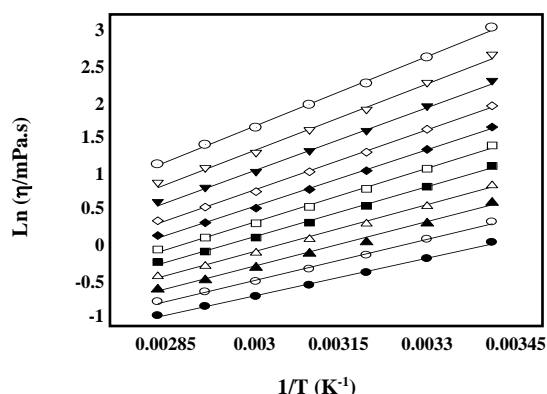


Fig. 1: Variation of the shear viscosity logarithm ($\ln(\eta)$) as a function of the inverse absolute temperature ($1/T$) for EG (1) + W (2) systems at some fixed mole composition x_1 . (Temperature domain = 293.15–353.15 K). (●): $x_1 = 0.000$; (○): $x_1 = 0.0312$; (▲): $x_1 = 0.0676$; (△): $x_1 = 0.1105$; (■): $x_1 = 0.1620$; (□): $x_1 = 0.2248$; (◆): $x_1 = 0.3031$; (◇): $x_1 = 0.4036$; (▼): $x_1 = 0.5370$; (▽): $x_1 = 0.7230$ and (⊙): $x_1 = 1.00$.

values of some nonelectrolyte solutions of binary system, equivalent to the Jones-Dole and Debye-Hückel equations [43,44] and the results reported previously [37-40] showed solute-solvent interaction feature at infinite dilution.

On the other hand, it is observed that the viscosity-absolute temperature dependence can be determined with an Arrhenius-type equation as follows:

$$\eta = (A_s) \cdot e^{\frac{E_a}{RT}} \quad (1)$$

Where E_a , R and A_s are, respectively, the activation energy, the perfect gas constant and the pre-exponential factor of the Arrhenius-type equation for the system. Under the logarithmic form, we can be rewritten the Eq. (1) as follows:

$$\ln \eta = \ln(A_s) + \frac{E_a}{RT} \quad (2)$$

Fig. 1 shows that the natural logarithm of shear viscosity $\ln(\eta)$ for which we plotted the dependence versus the inverse of absolute temperature ($1/T$) for EG (1) + W (2) fluid system in the whole domain of molar composition (x_1) is practically linear for absolute temperature over the studied absolute temperature domain (293.15 to 353.15) K, and the Arrhenius parameters E_a and A_s are thus independent of absolute

temperature. Using both least-squares fitting methods and graphics, $\ln(A_s)$ is the y-intercept and E_a/R is the slope of the straight line (Table 2) either for their corresponding systems ($x_1 \neq 0$) or for the pure constituents (i.e. at $x_1 = 0$ or $x_1 = 1$).

Correlation between Arrhenius parameters

Fig. 2 presents the variation of the entropic factor $-R \cdot \ln(A_s)$ (Pa.s) / (J/K.mol) and the activation energy E_a (kJ/mol) for {EG (1) and W (2)} fluid systems in the absolute temperature domain (293.15 to 353.15) K. We observe a strict increasing of the Arrhenius activation energy $E_a(x_1)$ of shear viscosity versus the logarithm of the Arrhenius entropic factor $-R \cdot \ln(A_s)$. Since, the A_s -values are closely related to the shear viscosity of the system in vapor-phase (Table 1, Fig. 3) [5], which it is greater, in W-rich region, than of EG and we can suppose that the W molecules bounding are more correlated and ordered in vapor state. This pre-exponential factor (A_s) is closely related to the shear viscosity of the same system in vapor-phase, it is equivalent to the shear viscosity at infinite absolute temperature ($A_s = \eta_\infty$) at the normal boiling point and at the same used pressure (Table 1, Fig. 3). Moreover, for some other before studied binary fluid systems, the correlation curve of these two quantities shows roughly linear behavior, we can cite for example water with isobutyric acid (IBA) [8,40], 1,4-dioxane [5] and N,N-dimethylacetamide with 2-ethoxyethanol (EOE) [1], water [2], formamide (FA) [4], N,N-dimethylformamide (DMF) [6], 2-methoxyethanol (MOE) [9] and methanol (Met) [7], and DMF with (Met) [3], 1,4-dioxane [10] and 2-propanol [11] and finally 1,2-dimethoxyethane (DME) with propylene carbonate (PPC) [12]. We note that for each binary fluid system; even in the case of the existing of different domains with distinct behaviors (existence of singular points) [1-13], we remark that all different branches exhibit almost the same slope of their quasi-straight line where it we can write the following standardized empirical expression at a reliable approximation:

$$-\frac{E_a}{R} = T_A \cdot \ln(A_s) + B \quad (3)$$

where the constant B is the y-intercept when $\ln(A_s)$ is mathematically null and it is thoroughly related to the shear viscosity of the binary fluid system at boiling point; while each binary fluid system is characterized by an x-intercept $1/T_A$ and is analog to a reciprocal of an absolute

Table 1. Arrhenius activation energy E_a (kJ/mol), Arrhenius pre-exponential factor $A_s / (10^{-6} \text{ Pa}\cdot\text{s})$, the entropic factor of Arrhenius $-R \ln(A_s/\text{Pa}\cdot\text{s})$ (J/K.mol) and, enthalpy and entropy of activation of viscous flow ΔH^* (kJ/mol) and ΔS^* (J/K.mol) for {EG (1) + W (2)} systems as a function of the mole composition (x_1) of EG over the temperature domain (293.15 to 353.15) K.

| x_1 | E_a | A_s | $-R \ln A_s$ | ΔH^* | ΔS^* |
|--------|--------|-----------------------------|--------------|--------------|--------------|
| | kJ/mol | $\mu\text{Pa}\cdot\text{s}$ | J/K.mol | kJ/mol | J/K.mol |
| 0.0000 | 14.801 | 2.2550 | 108.11 | 14.417 | 17.693 |
| 0.0312 | 16.166 | 1.7135 | 110.39 | 15.672 | 19.092 |
| 0.0676 | 17.420 | 1.3385 | 112.44 | 16.851 | 20.331 |
| 0.1105 | 18.439 | 1.1599 | 114.21 | 17.767 | 21.406 |
| 0.1620 | 19.683 | 0.91279 | 116.29 | 18.962 | 22.567 |
| 0.2248 | 21.130 | 0.65237 | 118.42 | 20.429 | 23.809 |
| 0.3031 | 22.242 | 0.54496 | 119.92 | 21.517 | 24.534 |
| 0.4036 | 23.548 | 0.43014 | 121.88 | 22.808 | 25.124 |
| 0.5370 | 24.753 | 0.36500 | 123.25 | 24.007 | 25.400 |
| 0.7230 | 26.117 | 0.29679 | 124.97 | 25.387 | 25.744 |
| 1.0000 | 27.393 | 0.26344 | 125.96 | 26.685 | 25.047 |

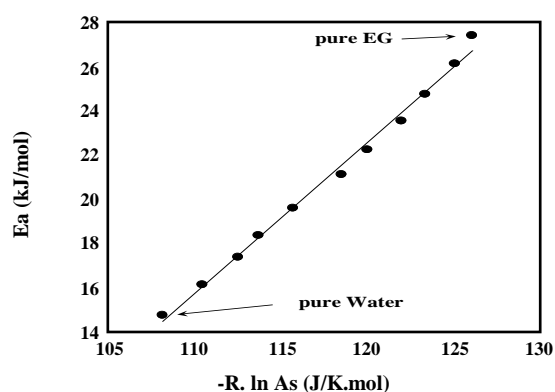


Fig. 2: Variation of the Arrhenius activation energy E_a (kJ/mol) of shear viscosity and the logarithm of the entropic factor of Arrhenius $-R \ln(A_s/\text{Pa}\cdot\text{s})$ / (J/K.mol) for {EG (1) and W (2)} systems. (Temperature domain 293.15 – 353.15 K).

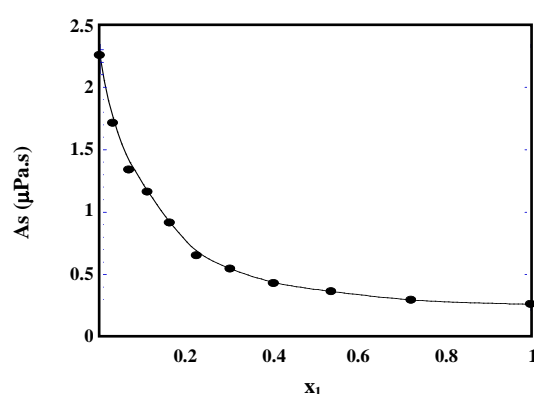


Fig. 3: Variation of the infinite-temperature viscosity (η_∞ or A_s) in $\mu\text{Pa}\cdot\text{s}$ for EG (1) + W (2) systems as a function of the mole composition x_1 of EG. (Temperature domain: 293.15 – 353.15 K).

temperature, where this is called Arrhenius viscosity temperature for the corresponding binary fluid system. Remarkable, we observe that for all the studied systems the T_A -values are rather correlated with the boiling points (T_{bi}) of the corresponding constituting pure fluid constituents or narrowly related to the vaporization temperature in the isobaric diagram of the corresponding binary fluid system for different molar compositions. The average T_A value

in the case of the EG-W systems is equal to 681.9 K with correlation factor 0.99701, whereas the boiling point of W and EG are 373.15 K and 466.45 K respectively; As concluding statement it may be observed that that the viscosity Arrhenius temperature T_A is greater than the boiling points (T_{bi}) which indicates that in the isobaric liquid vapor equilibrium of the binary fluid system EG-W there exists a probably convexity in the shape of the T - x_1 curve.

THERMODYNAMIC CHARACTERS

Enthalpy of activation of viscous flow

If we add the absolute reaction rate theory of *Aliet al.* [45] and *Eyring et al.* [46] for the fluid phase, the viscous and the free energy (ΔG^*) of activation of viscous flow are related as follows.:

$$\Delta G^*(x_1, T, P) = RT \cdot \ln \left(\frac{\eta(x_1, T, P) \cdot V(x_1, T, P)}{h N_A} \right) \quad (4)$$

Where R , η , h , N_A and V are the perfect gas constant, shear viscosity of binary fluid system, Plank's constant,

Avogadro's number and molar volume of fluid system (Eq. (6)), at molar composition (x_1), absolute temperature T and pressure P respectively, and:

$$\Delta G^* = \Delta H^* - T\Delta S^* \quad (5)$$

$$V = \frac{x_1 M_1 + (1 - x_1) M_2}{\rho} \quad (6)$$

Where ΔH^* and ΔS^* are the entropy and the enthalpy of activation of viscous flow, M_1 and M_2 are the molecular weight of the pure fluid constituent of EG and W respectively and ρ is the density of binary fluid system. By assuming that the parameters of activation of viscous flow ΔS^* and ΔH^* [45,46] are practically independent of absolute temperature, the temperature dependence of (ΔG^*) leads us to determine graphically the entropy ΔS^* of activation of viscous flow and the enthalpy ΔH^* which are given in Table 1. So, in Fig. 4, the ratio ($\Delta G^*/T$) against $1/T$ is represented in the studied domain of absolute temperature from 293.15 to 353.15 K for the fluid system of EG (1)+W (2) systems at some representative mole compositions of EG, the result clearly shows a linear behavior with positive slope. Using both, graphical method and least-square fit, the y -intercept is equal to ($-\Delta S^*$) and the slope is equal to ΔH^* .

Analysis of the enthalpy (ΔH^*) of activation of viscous flow and those of the Ea -values in the Fig. 5 shows that the Ea and ΔH^* values are very closely related. Hence, we can express the increment ΔH^* as the difference between Ea -values and those of (ΔH^*) as follow:

$$\delta H^* = Ea - \Delta H^* \quad (7)$$

and:

$$\delta H^* = x_1 \delta H_1^* + x_2 \delta H_2^* + x_1 x_2 \delta H_{12}^* \quad (8)$$

Where $\Delta H^*_{12} = 0.807$ kJ/mol is an interaction increment, $\Delta H^*_2 = 0.480$ kJ/mol and $\Delta H^*_1 = 0.673$ kJ/mol are enthalpy increments related to the pure constituents W (2) and EG (1) respectively [14,18,22].

In consideration of some thermodynamic Maxwell relations (Eqs. (9), (10) and (11)) and the eqs 2, 4, 5 and 6, we can see that the isobaric thermal expansion coefficient (α) of the fluid system is related to the enthalpy increment (ΔH^*) which is expressed by (Eq.(12)). Furthermore, this enthalpy increment is equal to the difference between the two activation energies related to the shear and kinematic viscosity respectively.

$$\alpha(x_1, T, P) = \frac{1}{V} \cdot \left(\frac{\partial V(x_1, T)}{\partial T} \right)_P = -\frac{1}{\rho} \cdot \left(\frac{\partial \rho(x_1, T)}{\partial T} \right)_P \quad (9)$$

$$\left(\frac{\partial V(x_1, T)}{\partial T} \right)_P = - \left(\frac{\partial S(x_1, T)}{\partial P} \right)_T \quad (10)$$

$$\Delta H^* = \left(\frac{\partial (\Delta G^*/T)}{\partial (1/T)} \right)_P \quad (11)$$

$$Ea - \Delta H^* = R\alpha T^2 \quad (12)$$

with T is approximately the average values over the studied domain of absolute temperature. The Eq.(12) is with a good accord with data given from literature [14,18,22] studied volumetric quantities of EG-W systems.

In future work, we will be interested in modeling the data of some thermophysical and thermodynamic quantities of EG-W binary system with and without presence of nanoparticles in order to inject them in eventual simulation programs. For this purpose, we preliminarily propose here the following empirical relations (Eqs.(13) and (14)) which are in excellent agreement with our experimental data in the studied absolute temperature domain.

$$Ea = \frac{\left(\frac{Ea_1}{M_2} - \frac{Ea_2}{M_1} \right) x_1 + \frac{Ea_2}{M_1}}{\left(\frac{1}{M_2} - \frac{1}{M_1} \right) x_1 + \frac{1}{M_1}} \quad (13)$$

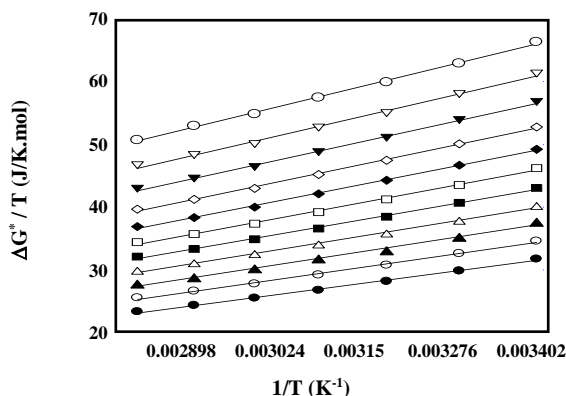


Fig. 4: Variation of $\Delta G^*/T$ as a function of the inverse of absolute temperature ($1/T$) for EG (1) + W (2) systems at some fixed mole composition x_1 . (Temperature domain = 293.15 – 353.15 K). (●): $x_1 = 0.000$; (○): $x_1 = 0.0312$; (▲): $x_1 = 0.0676$; (△): $x_1 = 0.1105$; (■): $x_1 = 0.1620$; (□): $x_1 = 0.2248$; (◆): $x_1 = 0.3031$; (◇): $x_1 = 0.4036$; (▼): $x_1 = 0.5370$; (▽): $x_1 = 0.7230$ and (⊙): $x_1 = 1.00$.

$$\Delta H^* = \frac{\left(\frac{\Delta H_1^*}{M_2} - \frac{\Delta H_2^*}{M_1}\right)x_1 + \frac{\Delta H_2^*}{M_1}}{\left(\frac{1}{M_2} - \frac{1}{M_1}\right)x_1 + \frac{1}{M_1}} \quad (14)$$

where the indexes 1 and 2 designate the pure constituents EG(1) and W(2) respectively. The corresponding values of (Ea_i) and (ΔH_i^*) are given from Table 1 at $x_1 = 1$ and $x_1 = 0$ for $i = 1$ and 0, respectively.

Furthermore, the variation of ΔS^* and $-R \cdot \ln(As)$ with the mole composition of EG (x_1) are plotted in Fig. 6. It is important to note that compared with Fig. 3 we cannot significantly inspire a change in curvature and the profile of the two parameters ($-R \cdot \ln(As)$ and Ea) increasing monotonically as the mole composition of EG varies, suggests that the solvent's structure is more ordered in its present form and in the vapor phase than the other constituent.

The difference between the Arrhenius entropic factor ($-R \cdot \ln(As)$) and the entropy of activation of viscous flow (ΔS^*) can be expressed by an entropic increment (δS^*) as follows:

$$\delta S^* = -R \cdot \ln(As) - \Delta S^* \quad (15)$$

and:

$$\delta S^* = x_1 \cdot \delta S_1^* + x_2 \cdot \delta S_2^* + x_1 x_2 \cdot \delta S_{12}^* \quad (16)$$

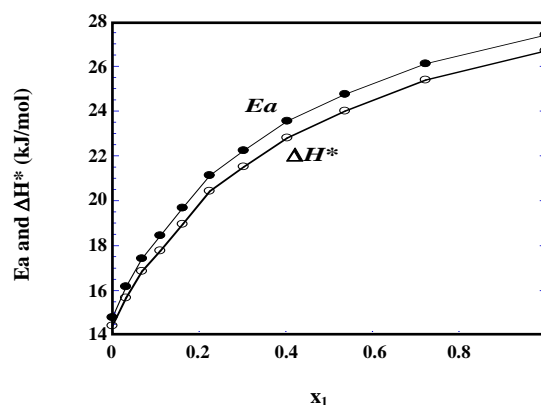


Fig. 5: Variation of Arrhenius activation energy Ea and enthalpy of activation of viscous flow ΔH^* for EG (1) + W (2) systems as a function of the mole composition x_1 of EG calculated. (Temperature domain = 293.15 – 353.15 K). (●): Ea (kJ/mol); (○): ΔH^* (kJ/mol).

where $\delta S_{12}^* = 7.64$ J/K.molis an interaction increment (Fig. 6), $\delta S_1^* = 100.7$ J/K.mol and $\delta S_2^* = 90.8$ J/K.mol are entropy increments related to the pure constituents EG (1) and W (2) respectively. In like manner and considering the Eq.(17) and the precedent thermodynamic Maxwell relations, we can deduce that the entropy increment (δS^*) is related to the isobaric thermal expansion coefficient-temperature product at constant pressure and the logarithm of molar volume of the system (Eq.(18)).

$$\Delta S^* = -\left(\frac{\partial(\Delta G^*)}{\partial(T)}\right)_p \quad (17)$$

$$\delta S^* = R \cdot \left[\ln\left(\frac{V(x_1, T, P)}{hN_A}\right) + \alpha \cdot T \right] \quad (18)$$

Partial molar activation energy

Notice that the close correlation (eqs 7 – 18) for the thermodynamic functions of activation for viscous flow of binary liquid systems, i.e. ΔH^* , Ea , ΔS^* and $-R \cdot \ln(As)$ discussed in previous works [1-13] and described in the (*Enthalpy of activation of viscous flow*)-section, we can admit that $Ea(x_1)$ is approximately a thermodynamic function and consequently we can establish the partial molar quantities Y_1 and Y_2 for EG (1) and W (2) respectively via the following two equations:

$$Y_{1,T,P}(x_1) = Y_{T,P}(x_1) + (1 - x_1) \cdot \left(\frac{\partial Y(x_1)}{\partial(x_1)}\right)_{T,P} \quad (19)$$

Table 2: Arrhenius' current temperatures (T_{Ai}) / K (eq (21)), partial molar quantities relative to the activation energies Ea_i (kJ/mol) and the logarithm of the entropic factor of Arrhenius $-R \cdot \ln(As_i / Pa \cdot s)$ / ($J \cdot K^{-1} \cdot mol^{-1}$) for {EG (1) + W (2)} systems as a function of the mole composition of EG (x_1) over the absolute temperature domain (393.15 to 353.15) K.

| x_1 | T_{Ai} | Ea_1 | Ea_2 | $(-R \cdot \ln As)_1$ | $(-R \cdot \ln As)_2$ |
|--------|----------|--------|--------|-----------------------|-----------------------|
| | K | kJ/mol | kJ/mol | J/K.mol | J/K.mol |
| 0.0000 | 748.29 | 55.639 | 14.801 | 178.17 | 108.11 |
| 0.0312 | 753.33 | 51.149 | 15.039 | 169.74 | 108.48 |
| 0.0676 | 771.22 | 46.631 | 15.303 | 161.20 | 108.91 |
| 0.1105 | 790.51 | 42.091 | 15.500 | 152.77 | 109.42 |
| 0.1620 | 816.80 | 38.125 | 16.117 | 145.33 | 110.68 |
| 0.2248 | 853.31 | 34.894 | 17.138 | 139.01 | 112.45 |
| 0.3031 | 891.62 | 32.015 | 17.991 | 133.55 | 113.99 |
| 0.4036 | 971.00 | 30.077 | 19.130 | 130.30 | 116.19 |
| 0.5370 | 1055.4 | 28.653 | 20.230 | 128.05 | 117.68 |
| 0.7230 | 1210.8 | 27.733 | 21.897 | 126.90 | 119.92 |
| 1.0000 | 1332.3 | 27.393 | 22.887 | 125.96 | 125.96 |

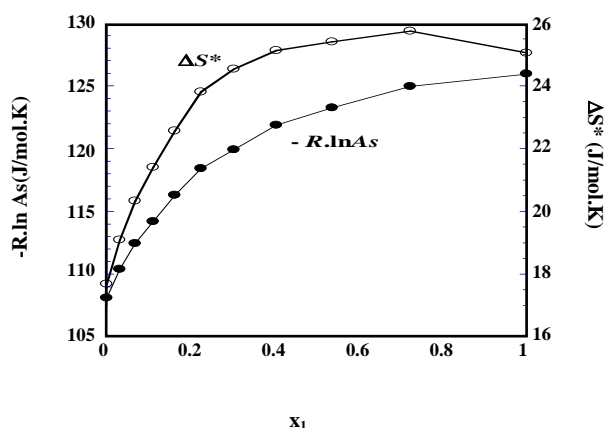


Fig. 6: Variation of the logarithm of the entropic factor of Arrhenius $-R \cdot \ln(As)$ ($J K^{-1} mol^{-1}$) and entropy of activation of viscous flow ΔS^* ($J K^{-1} mol^{-1}$) for EG (1) + W (2) systems as a function of the mole composition x_1 of EG. (Temperature domain = 293.15–353.15 K). (●): $-R \cdot \ln(As/Pa \cdot s)$ / ($J/K.mol$) ; (○): ΔS^* (kJ/mol).

$$Y_{2,T,P}(x_1) = Y_{T,P}(x_1) - x_1 \cdot \left(\frac{\partial Y(x_1)}{\partial (x_1)} \right)_{T,P} \quad (20)$$

$Y_{i,T,P}(x_1)$ is be regarded as the partial molar quantity relative to the entropic factor of Arrhenius $-R \cdot \ln(As_i)$ or the partial molar activation energy Ea_i .

Ensuing individual contribution of every pure constituent (i) and preferential phenomena in concurrence

of every fixed binary system composition (x_1, x_2) is accomplished by the values partial molar activation energy $Ea_i(x_1)$, that represents a very interesting approximation.

Table 2 shows the values of $-R \cdot \ln(As_1)$, $-R \cdot \ln(As_2)$, $Ea_1(x_1)$ and $Ea_2(x_1)$, and the Fig. 7 presents a prediction of the variation of $Ea_1(x_1)$ and $Ea_2(x_1)$ against mole composition of EG (x_1). The partial molar activation energy $Ea_1(x_1)$ of EG monotonously decreases from a maximum high positive value (about 55.64 kJ/mol), then decreases rapidly to reach a minimum at ($x_1 \approx 1$, pure EG) (about 27.39 kJ/mol). Additionally, the partial molar activation energy $Ea_2(x_1)$ of W increases very slightly from a minimum positive value (about 14.80 kJ/mol) to reach a maximum value at infinite dilution ($x_2 \approx 0$) (about 22.89 kJ/mol).

The hypothesis of the ease of integration of the introduced molecules of W in the EG-rich region explains the monotonic variation and this remains valid for the inverse cases. Also, the activation energy Ea varies in a strictly monotonic manner, which does not reflect a considerable change in the structure of the solvent when we insert one constituent into the other in the systems.

Nonetheless, if we eliminate the hypothesis of Eq. (3) that the Arrhenius temperature (T_A) nothing is more constant over the whole domain of composition, we can review it as variable Arrhenius' current temperatures (T_{Ai}).

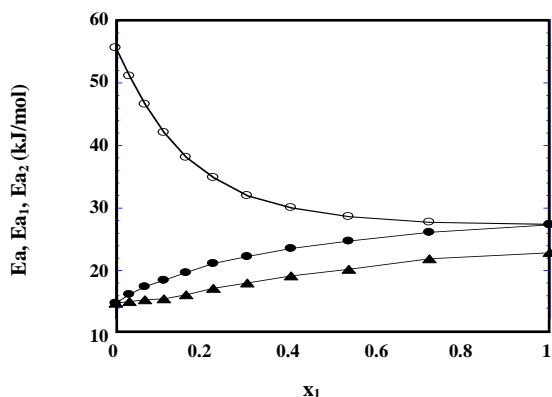


Fig. 7: Variation of Arrhenius activation energy Ea /(kJ/mol) and partial molar activation energies of shear viscosity (Eqs. (19) and (20)) Ea_i /($\text{kJ}\cdot\text{mol}^{-1}$) for {EG (1) + W (2)} systems as a function of the mole composition of EG (x_1). (Temperature domain = 293.15 – 353.15 K). (●): $Ea(x_1)$; (○): $Ea_1(x_1)$ and; (▲): $Ea_2(x_1)$.

It can be determined from the following partial derivatives at selected molar composition (x_1):

$$T_{Ai} = \left(\frac{\partial(Ea_i)}{\partial(-R \cdot \ln(As_i))} \right)_P \quad (21)$$

We notice that the form of eq (21) is justified from the fact of the behavior's analogy of the Arrhenius parameters $-R \cdot \ln(As)$ and $Ea(x_1)$ with the entropy (ΔS^*) of activation of viscous flow and the enthalpy (ΔH^*) respectively ([1-13], Figs. 5 and 6, Eqs. (7) to (18)). Indeed, considering the partial derivatives functions of Maxwell equations and the Gibbs free energy expression (Eq. (5)), the Eq. (21)

can be considered to be equivalent to the partial derivative with respect to (ΔS^*) at constant pressure.

$$\left(\frac{\partial(\Delta H^*)}{\partial(\Delta S^*)} \right)_P = T \quad (22)$$

Hence, the slope of the tangent to a selected point of the graph represents the experimental values of T_{Ai} (i.e. at selected system composition (x_1)). The most striking result has been obtained particularly in the vicinity of the two limits of composition domain as application of this definition. Hence, from the slopes of the two curves given in Figs. 8 and 9 we obtain interesting limiting values of Arrhenius' current temperatures (T_{Ai}) (see dashed lines) such as: for a very high concentration of one constituent

(i) we get that $T_{cA1}(x_1 \approx 1) = 475.6 \text{ K} \approx T_{b1}$ for the first curve ($i=1$), $T_{cA2}(x_2 \approx 1) = 392.7 \text{ K} \approx T_{b2}$ for the second curve ($i=2$), whereas for a very high dilution of the constituent (i) in the second one (j), we get that $T_{cA1}(x_1 \approx 0) = 597.5 \text{ K}$ for the first curve, and, $T_{cA2}(x_2 \approx 0) = 227.5 \text{ K}$ for the second curve. With this find, we can predict the boiling point of the two pure constituents at constant pressure per investigation of the shear viscosity-temperature dependence in the corresponding liquid phase. Notice that these significant results are practically verified for other studied anterior works [1–13] in different binary fluid systems (Table 3). Based on the accuracy of data and the errors made in the computations, we can conclude that the current Arrhenius temperature (T_{Ai}) is very nearly to its corresponding boiling point for a high concentration of one constituent ($x_i \approx 1$) whereas in the other cases, a strong correlation exists between the Arrhenius' current temperatures (T_{Ai}) at ($x_i \neq 1$) and the vaporization temperature of the isobaric liquid vapor equilibrium of the corresponding binary system.

The following equation asserts all these observations:

$$\lim_{x_i \rightarrow 1} \left(\frac{\partial(Ea_i)}{\partial \left(\frac{\partial(\ln As_i)}{\partial x_i} \right)} \right) \approx -R \cdot T_{bi} \quad (23)$$

We can then estimate with a good approximation, the boiling point (T_{bi}) of the pure constituent (i).

The present Arrhenius temperatures (T_{cAi})/K at ($x_i \approx 1$) and the corresponding boiling point T_{bi} /K for the pure component (i) are correlated by the couple of constituents from some binary fluid systems appeared in anterior works (Eqs. (21) or (23)) [1-13] given in Table 3 and plotted in Fig. 10.

In order to do illustrate of the linear least square fit, we applied the methods for linear-least-squares fitting ($Y = -29.41 + 1.07X$); with a correlation coefficient $R = 0.96801$ is close to the straight line corresponding to the first bisector ($Y = +X$), where $Y = T_{cAi}$ and $X = T_{bi}$. Indeed, the slope which deviates from the unit to about 6.9% is equal to 1.069 and the absolute value of the y-intercept (29.408) is less than the standard deviation ($\sigma = 46.76$) related to the ensemble of boiling points used in this correlation (414.71 K is the mean value of T_{bi} values).

Starting from the fact for the mutual correlations between the current Arrhenius temperatures (T_{Ai}) defined by the ratio of the two Arrhenius parameters some pure fluids [1–13] and the corresponding boiling point (T_{bi}), the Fig. 11 shows a strong causal correlation which

Table 3: Comparison between the current Arrhenius temperature (T_{A_i}) / K (eq 23) for ($x_i \approx 1$) and the corresponding boiling point T_{b_i} / K of the pure constituent (i) in some binary systems.

| Constituent 1 | Constituent 2 | Ref. | T_{A_i} / K | T_{b_i} / K | T_{A_2} / K | T_{b_2} / K |
|---------------|---------------|------|---------------|---------------|---------------|---------------|
| DMF | Dioxane | [10] | 400.7 | 426.15 | 325.1 | 374.25 |
| DMF | 2-Propanol | [11] | 398.3 | 426.15 | 367.6 | 355.15 |
| DMF | Met | [3] | 394.5 | 426.15 | 316.3 | 337.85 |
| DMA | Met | [7] | 418.7 | 438.15 | 319.4 | 337.85 |
| DMA | FA | [4] | 429.1 | 438.15 | 479.6 | 483.65 |
| DMA | EOE | [1] | 440.0 | 438.45 | 410.0 | 408.15 |
| DMA | DMF | [6] | 330.6 | 438.15 | 329.1 | 426.15 |
| DMA | MOE | [9] | 361.5 | 438.15 | 332.3 | 397.65 |
| DMA | Water | [2] | 456.1 | 438.45 | 378.5 | 373.15 |
| Dioxane | Water | [5] | 378.6 | 374.25 | 380.5 | 373.15 |
| IBA | Water | [8] | 433.1 | 428.15 | 362.9 | 373.15 |
| DME | PPC | [12] | 361.1 | 358.15 | 530.9 | 515.15 |
| EG | Water | a | 475.6 | 466.45 | 392.7 | 373.15 |

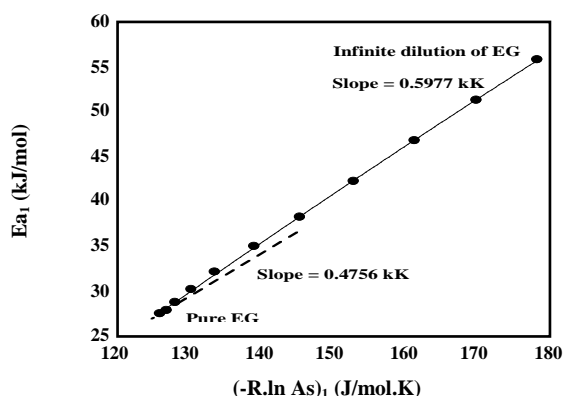


Fig. 8: Correlation between the partial molar quantities relative to the activation energies E_{a1} (kJ/mol) and the logarithm of the entropic factors of Arrhenius $-R \ln(As_1/Pa \cdot s)$ (J/mol.K) related to EG for {EG (1) + W (2)} systems. (Temperature domain = 293.15–353.15 K).

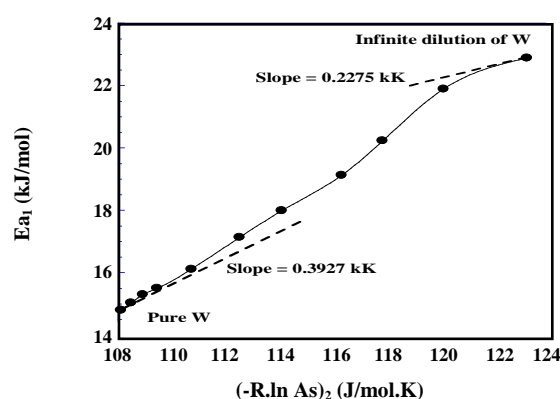


Fig. 9: Correlation between the partial molar quantities relative to the activation energies E_{a2} (kJ/mol) and the logarithm of the entropic factors of Arrhenius $-R \ln(As_2/Pa \cdot s)$ (J/mol.K) related to W for {EG (1) + W (2)} systems. (Temperature domain = 293.15–353.15 K).

can be expressed empirically and mathematically by a hyperbolic or homographic dependence (i.e. with vertical and horizontal asymptotes) as follows:

$$E_a = \frac{R \cdot \omega}{\left(\frac{1}{T_A} - \frac{1}{T_b}\right) - \frac{1}{\tau}} + \varepsilon_0 \quad (24)$$

Where ω is dimensionless constant, R is the perfect gas constant, τ is equivalent to an absolute temperature

statistically related to the minimal values that the difference between the reciprocal of Arrhenius and boiling points can be theoretically taken for the set of treated liquids organic group and ε_0 is equivalent to the minimal value that the activation energy can be theoretically taken for the set of studied of viscous fluids.

Also, A new equation suggest by Haj-Kacem[47-49] presents for moderate and low viscous pure fluids

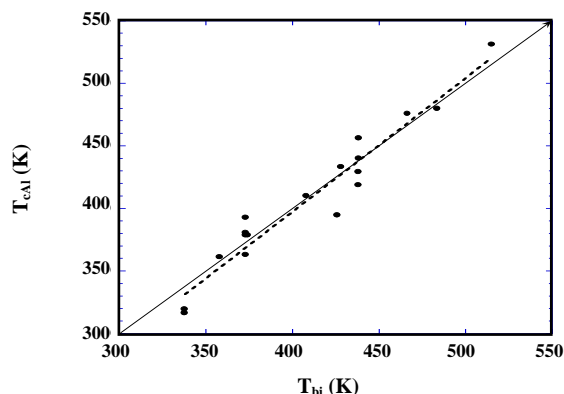


Fig. 10: Correlation between the current Arrhenius temperatures (T_{Ai}) / K for ($x_i \approx 1$) for the pure constituent (i) and the corresponding boiling point T_{bi} / K determined by couple of constituents from some binary fluid systems studied at previous works (eqs (21) and (23)) [1-13]. (—): the first bisector ($Y = X$) in the present axis system and (---): linear least square fit.

interesting causal correlation with good concordance between the two viscosity Arrhenius parameters ($\ln A_s$ and E_a) (eq (25)) which have not very low values of pre-experimental factor ($42 \text{ nPa}\cdot\text{s} < A_s < 45 \text{ }\mu\text{Pa}\cdot\text{s}$, i.e. $-17 < \ln(A_s / \text{Pa}\cdot\text{s}) \leq 10$) and not very high values of activation energy ($6 < E_a < 30 \text{ kJ}\cdot\text{mol}^{-1}$).

Also, a new equation suggest by Haj-Kacem[47-49] presented for moderate and low viscous pure fluids correlating ($\ln A_s$) and (E_a) (viscosity Arrhenius parameters) (Eq. (25)) with good accordance that have not very low values of pre-experimental factor ($42 \text{ nPa}\cdot\text{s} < A_s < 45 \text{ }\mu\text{Pa}\cdot\text{s}$, i.e. $-17 < \ln(A_s / \text{Pa}\cdot\text{s}) \leq 10$) and not very high values of activation energy ($6 < E_a < 30 \text{ kJ}\cdot\text{mol}^{-1}$).

$$E_a = \lambda \cdot R \times (-\ln A_s)^{\alpha_0} \quad (25)$$

where R is the perfect gas constant, $\alpha_0 = 2.933$ and $\lambda \approx 1 \text{ K}$.

We observe that this equation can be also used for the two parameters related to some binary fluid systems with a good approximation. Furthermore, this model would be interesting in various physicochemical areas. Also, it will be very advantageous for engineering data and it will let to estimate one unknown parameter when the second one is known (Eq. (25) or Eq. (26)), or can be otherwise estimate by some theories suggested in the literature [47].

$$\ln A_s = -\left(\frac{E_a}{\lambda R}\right)^{\frac{1}{\alpha_0}} \quad (26)$$

By means of a wide range of validation ($5 < E_a < 60 \text{ kJ/mol}$) and ($-25 < \ln(A_s / \text{Pa}\cdot\text{s}) < -9$) we can get the same results with the Messaâdi equation [50,51]:

$$\ln(A_s) = \frac{-E_a}{RT_0 \left[1 - e^{-\frac{E_a}{\alpha_0 RT_0}} \right]} \quad (27)$$

$$E_a = -\frac{RT_0 \cdot [\ln(A_s) + \alpha_0]}{\left[1 - e^{\frac{RT_0 \cdot \ln(A_s)}{\gamma_0}} \right]} \quad (28)$$

Where (T_0) is the limiting Arrhenius temperature ($T_0 = 289 \pm 4$) K, R is the perfect gas constant, ($\gamma_0 = (37.5 \pm 5.6) \times 10^3$) and ($\alpha_0 = 9.02 \pm 0.54$) are two dimensionless constants[50,51].

In consideration of the causal correlations shown in Figs. 11 and 12, and eqs (24), (25) and (26) we can propose direct correlation between the activation energy E_a expressed and the boiling point T_b by the following empirical equation:

$$T_b(E_a) = -\frac{E_a}{68 - 4.05 \times E_a^{0.34}} \quad (29)$$

Nevertheless, this equation becomes invalid for the case of fluids with low or high shear viscosity notably for low boiling point ($T_b < T_{b, \text{Water}}$) due to the limitation of the validity domain of Eqs. (25) and (26) ($6 < E_a < 30 \text{ kJ/mol}$).

In the same way and in place of the activation energy (E_a) variable, we can obtain the following empirical expression eq (30) instead of eq (29) if we consider the ($\ln A_s$) as a variable:

$$T_b(\ln A_s) = \frac{(-\ln A_s)^{2.933}}{8.2 + \ln A_s} \quad (30)$$

In addition, due to the entropic factor domain restriction ($-17 < \ln(A_s / \text{Pa}\cdot\text{s}) < -10$) we mark the same previous conclusion. Although, as was expected from the Fig. 12 presenting the comparison of experimental and calculated boiling points by means of the two Eqs. (29) and (30), we can conclude that there are also two classes of causal correlations. Indeed, we indicate that the two different points' forms don't match.

We conclude that the causal correlation, between the boiling point and the two Arrhenius parameters (E_a) or ($\ln A_s$) is implicitly due to varied chemical and physical quantities for which there are some that are common for the two Arrhenius parameters although others are related only to a single parameter (E_a) or ($\ln A_s$). Explicit expression of $T_b(E_a, \ln A_s)$ will be proposed in future work

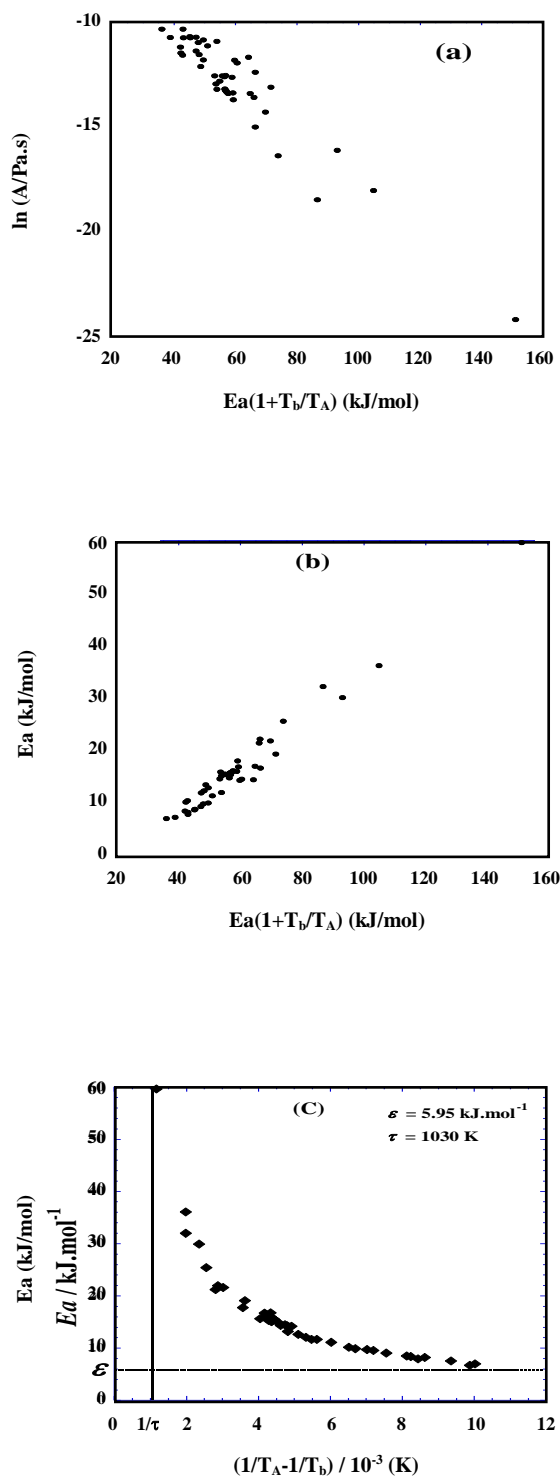


Fig. 11: Correlations between Arrhenius parameters (E_a or $\ln(A_s)$) and transformed variables as a function of $(1/T_A$ and $1/T_b)$ of some pure fluids [1-13].

to correcting this observation, for which the two viscosity Arrhenius parameters appear alternatively in the composition [52-55].

CONCLUSIONS

Some novel theoretical approaches have been released by studying the derived partial molar quantities and variation of Arrhenius activation energy against molar composition, on the basis of the same experimental data of densities and shear viscosities of EG + W binary fluid systems at atmospheric pressure and for seven value of absolute temperature subject to a range 293.15-353.15 K [14].

Variation of the viscosity's Arrhenius' activation energy and the enthalpy of activation of viscous flow of pure constituents (EG and W) as a function of the system molar composition is predicted. For binary fluid systems, the correlation between the two Arrhenius parameters ($\ln(A_s)$) and (E_a) makes it possible to predict the viscosity Arrhenius temperature which characterizes the binary fluid system and can supply information on the vaporization temperature of the isobaric liquid-vapor equilibrium.

So, we have determined the partial molar activation energy in order to understand the contributions of every pure constituent in the system assuming that the activation energy is a thermodynamic quantity. Over the studied domain of absolute temperature, the correlation between the logarithm of the entropic factors of Arrhenius for EG + W systems and the molar quantities relative to the activation energies can show an approximately linear behavior i.e. no remarkable change in curvature. An empirical equation is then suggested at the base of the quasi-linear behavior introducing a new parameter T_A which characterizes every binary system and is denoted as the viscosity Arrhenius temperature. In the case of molar quantities, over the whole domain of composition we consider that the Arrhenius temperature (T_A) is not any more a constant, then we introduce for every pure constituent (i) a new concept of the Arrhenius' current temperature (T_{Ai}) to find its value at the two extreme positions i.e., at very high concentration and very high dilution respectively.

The obtained results of the studied binary system give an important fact that the isobaric boiling point (T_{bi}) of the pure constituents is strongly dependent or very close to the viscosity Arrhenius' current temperature (T_{Ai}).

Moreover, two empirical equations are proposed as a consequence of the causal correlation and from which we can predict reliable value of the boiling point values using

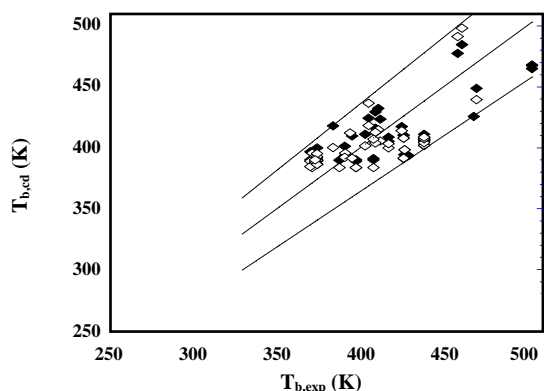


Fig. 12: Correlation between the experimental boiling point $T_{b,exp}$ / K and the calculated boiling point ($T_{b,cal}$) / K for some pure fluids studied at previous work [47]; (◆): from eq (29) and (◇): from eq (30). (—): around the first bisector ($Y = +X$ and $Y = (1.00 \pm 0.09) \times X$) in the present axis system.

only the viscosity Arrhenius parameters. Finally, with more mathematical manipulations we can confirm that we will be capable to disclose some physical meanings of the viscosity Arrhenius parameters which can be definitely develop as well as improve the thermodynamic theories and also to predict some information on liquid-vapor diagrams by studying the shear viscosity with a view to absolute temperature and composition only in the liquid phase of binary systems. In the same context, we have suggested new original empirical equation predicting the boiling point for some pure liquid solvents through one viscosity Arrhenius parameter such as (E_a) or $\ln(A_s)$.

Moreover, a supplementary study on the eventual relationship between the physicochemical quantities of numerous binary fluid systems and the Arrhenius temperature can prove from this method an evidence of the ability to predict the quantities of other untreated fluid systems [47-55]. In future to estimate the Arrhenius temperature and to prove the utility of such parameters, different systems will be studied to make this protocol most clear. To the best of our knowledge, there is not yet a predictive model for our initial hypothesis nor stronger theoretical and physical foundations of this study and we cannot supply our verifications clearly. We hope that these interesting and novel experimental results can be well exploited by researchers to elaborate new theoretical approaches. Likewise, the rate of shear viscosity variation due to the changes in pressure is considered a priority question in future prospects, notably the correlation that can be deduced from the theories previously available.

Finally, by using the concept of Arrhenius temperature and mathematical techniques, we have suggested reliable estimation of the boiling temperature through the viscosity-temperature dependence and proposed interesting relationship between the two principal Arrhenius parameters for which in absence of experimental data, we can predict reliable value of these physicochemical parameters for eventual simulation in fluid engineering, especially for nanofluids involved in EG systems.

Disclosure statement

No potential conflict of interest was reported by the authors.

Received :Dec. 31, 2018 ; Accepted :Mar. 11, 2019

REFERENCES

- [1] Hichri M., Das D., Messaâdi A., Bel Hadj Hmida E.S., Ouerfelli N., Khattech I., [Viscosity Arrhenius Activation Energy and Derived Partial Molar Properties in of N,N-dimethylacetamide + 2-Ethoxyethanol Binary Mixtures at Temperatures from 298.15 K to 318.15 K](#), *Phys. Chem. Liq.*, **51**:721–730 (2013).
- [2] Das D., Messaâdi A., Dhoubi N., Ouerfelli N., Hamzaoui A.H., [Viscosity Arrhenius Activation Energy and Derived Partial Molar Properties in N,N-dimethylacetamide + Water Binary Mixtures at Temperatures from 298.15 K to 318.15 K](#), *Phys. Chem. Liq.*, **51**:677–685 (2013).
- [3] Trabelsi Z., Dallel M., Salhi H., Das D., Al-Omair N.A., Ouerfelli N., [On the Viscosity Arrhenius Temperature for Methanol + N,N-dimethylformamide binary mixtures over the temperature range from 303.15 to 323.15 K](#), *Phys. Chem. Liq.*, **53**:529–552 (2015).
- [4] Dallel M., Das D., Bel Hadj Hmida E.S., Al-Omair N.A., Al-Arfaj A.A., Ouerfelli N., [Derived Partial Molar Properties Investigations of Viscosity Arrhenius Parameters in Formamide + N,N-dimethylacetamide Systems at Different Temperatures](#). *Phys. Chem. Liq.*, **52**:442–451 (2014).
- [5] Ouerfelli N., Barhoumi Z., Iulian O., [Viscosity Arrhenius Activation Energy and Derived Partial Molar Properties in 1,4-dioxane + Water Binary Mixtures from 293.15 K to 323.15 K](#), *J. Solution Chem.*, **41**:458–474 (2012).

- [6] Dhouibi N., Dallel M., Das D., Bouaziz M., Ouerfelli N., Hamzaoui A.H. [Notion of Viscosity Arrhenius Temperature for N,N-dimethylacetamide with N,N-Dimethylformamide Binary Mixtures and Its Pure Components](#), *Phys. Chem. Liq.*, **53**:275–292 (2015).
- [7] Salhi H., Dallel M., Trabelsi Z., Alzamel N.O., Alkhalidi M.A., Ouerfelli N., [Viscosity Arrhenius Activation Energy and Derived Partial Molar Properties in Methanol + N,N-dimethylacetamide Binary Mixtures the Temperatures from 298.15 K to 318.15 K](#). *Phys. Chem. Liq.*, **53**:117–137 (2015).
- [8] Das D., Salhi H., Dallel M., Trabelsi Z., Al-Arfaj A.A., Ouerfelli N., [Viscosity Arrhenius Activation Energy and Derived Partial Molar Properties in Isobutyric Acid + Water Binary Mixtures Near and Far Away from Critical Temperature from 302.15 K To 313.15 K](#), *J. Solution Chem.*, **44**:54–66 (2015).
- [9] Messaâdi A., Salhi H., Das D., Alzamel N.O., Alkhalidi M.A., Ouerfelli N., Hamzaoui A.H., [A Novel Approach to Discuss the Viscosity Arrhenius Behavior and to Derive the Partial Molar Properties in Binary Mixtures of N,N-Dimethylacetamide with 2-Methoxyethanol in the Temperature Interval \(from 298.15 To 318.15\) K](#), *Phys. Chem. Liq.*, **53**: 506–517 (2015).
- [10] Al-Omair N.A., Das D., Snoussi L., Sinha B., Pradhan R., Acharjee K., Saoudi K., Ouerfelli N., [A Partial Derivatives Approach for Estimation of the Viscosity Arrhenius Temperature in N,N-Dimethylformamide + 1,4-Dioxane Binary Fluid Mixtures at Temperatures from 298.15 K To 318.15 K](#), *Phys. Chem. Liq.*, **54**(5): 615–631 (2016).
- [11] Salhi H., Al-Omair N.A., Al-Arfaj A.A., Alkhalidi M.A., Alzamel N.O., Alqahtani K.Y., Ouerfelli N., [Correlation Between Boiling Temperature and Viscosity Arrhenius Activation Energy in N,N-Dimethylformamide + 2-Propanol Mixtures at 303.15 to 323.15 K](#), *Asian J. Chem.*, **28**(9):1972–1984 (2016).
- [12] Dallel M., Al-Zahrani A.A., Al-Shahrani H.M., Al-Enzi G.M., Snoussi L., Vranceanu N., Al-Omair N.A., Ouerfelli N., [Prediction of the Boiling Temperature of 1,2-Dimethoxyethane and Propylene Carbonate Through the Study of Viscosity Temperature Dependence of Corresponding Binary Liquid Mixtures](#), *Phys. Chem. Liq.*, **55**(4): 541–557 (2017).
- [13] Dallel M., Al-Arfaj A.A., Al-Omair N.A., Alkhalidi M.A., Alzamel N.O., Al-Zahrani A.A., Ouerfelli N., [A Novel Approach of Partial Derivatives to Estimate the Boiling Temperature via the Viscosity Arrhenius Behavior in N,N-dimethylformamide + Ethanol Fluid Systems](#), *Asian J. Chem.*, **29**(9):2038–2050 (2017).
- [14] Yang C., Ma P., Jing F., Tang D., [Excess Molar Volumes, Viscosities, and Heat Capacities for the Mixtures of Ethylene Glycol + Water from 273.15 K to 353.15 K](#), *J. Chem. Eng. Data*, **48**:836–840 (2003).
- [15] Han F., Zhang J., Chen G., Wei X., [Density, Viscosity, and Excess Properties for Aqueous Poly\(ethylene glycol\) Solutions from \(298.15 to 323.15\) K](#), *J. Chem. Eng. Data*, **53**(11):2598–2601 (2008).
- [16] Cristancho D.M., Delgado D.R., Martínez F., Fakhree M.A.A., Jouyban A., [Volumetric Properties of Glycerol + Water Mixtures at Several Temperatures and Correlation with the Jouyban-Acree Model](#), *Rev. Colomb. Cienc. Quím. Farm.*, **40**(1): 92–115, (2011).
- [17] Vural U.S., Muradoglu V., Vural S., [Excess Molar Volumes, and Refractive Index of Binary Mixtures of Glycerol + Methanol and Glycerol + Water at 298.15 K and 303.15 K](#), *Bull. Chem. Soc. Ethiopia.*, **25**(1):111–118 (2011).
- [18] Adam O.E.-A.A., Al-Dujaili A.H., Awwad A.M., [Volumetric Properties of Aqueous Solutions of Ethylene Glycols in the Temperature Range of 293.15–318.15 K](#), *ISRN Phys. Chem.*, Volume **2014**, Article ID 639813, 10 pages.
- [19] Nan Z., Liu B., Tan Z., [Calorimetric Investigation of Excess Molar Heat Capacities for Water + Ethylene Glycol from T = 273.15 to T = 373.15 K](#), *J. Chem. Thermodyn.*, **34**:915–926 (2002).
- [20] Nowak-Wózný D., Maczka T., [The DC Conduction Mechanism of Ethylene Glycol Water Solutions](#), *J. Elect. Eng.*, **58**(1): 55–57 (2007).
- [21] Kumar R.M., Baskar P., Balamurugan K., Das S., Subramanian V., [On the Perturbation of the H-Bonding Interaction in Ethylene Glycol Clusters upon Hydration](#), *J. Phys. Chem. A*, **116**:4239–4247 (2012).
- [22] Egorov G.I., Makarov D.M., Kolker A.M., [Volumetric Properties of the Water–Ethylene Glycol Mixtures in the Temperature Range 278–333.15 K at Atmospheric Pressure](#), *Russ. J. Gen. Chem.*, **80**(8):1577–1585 (2010).

- [23] Karimi A., Abdolahi Sadatlu M.A., Ashjaee M., Experimental Studies on the Viscosity of Fe Nanoparticles Dispersed in Ethylene Glycol and Water Mixture, *Therm. Sci.*, **20**(5):1661–1670 (2016).
- [24] Zhang J., Xiao J., Liu Y., Wei X., Solubility of Carbonyl Sulfide in Aqueous Solutions of Ethylene Glycol at Temperatures from (308.15 K to 323.15) K, *J. Chem. Eng. Data*, **55**:5350–5353 (2010).
- [25] Kulkarni D.P., Das D.K., Chukwu G.A., Temperature Dependent Rheological Property of Copper Oxide Nanoparticles Suspension (Nanofluid), *J. Nanosci. Nanotechnol.*, **6**(4):1–5 (2006).
- [26] Popa C.V., Nguyen C.T., Gherasim I., New Specific Heat Data for Al₂O₃ and CuO Nanoparticles in Suspension In Water and Ethylene Glycol, *Int. J. Therm. Sci.*, **111**:108–115 (2017).
- [27] Mahbubul I.M., Saidur R., Amalina M.A., Latest Developments on the Viscosity of Nanofluids, *Int. J. Heat Mass Transf.*, **55**:874–885 (2012).
- [28] Dehnavi M., Pazuki G., Vossoughi M., “Application of a Modified Excess Gibbs Energy Model for Correlating and Predicting the Viscosity of Nano Fluids”, *Proceedings of the 4th International Conference on Nanostructures (ICNS4)* 12-14 March, Kish Island, I.R. Iran (2012).
- [29] Kumar R.M., Baskar P., Balamurugan K., Das S., Subramanian V., Interaction of Ethylene Glycol–Water Clusters with Aromatic Surfaces, *RSC Adv.*, **3**:7798–7807 (2013).
- [30] Snoussi L., Chouikh R., Abid R., Simulation of Laminar Natural Convective Heat Transfer in a Three-Dimensional Rectangular Enclosure, *International Review of Mechanical Engineering, Int. Rev. Mech. Eng.*, **4**(3):297–304 (2010).
- [31] Snoussi L., Chouikh R., Ouerfelli N., Guizani A., Numerical Simulation of Heat Transfer Enhancement For Natural Convection in a Cubical Enclosure Filled With Al₂O₃/Water and Ag/Water Nanofluids, *Phys. Chem. Liq.*, **54**:703–716 (2016).
- [32] Snoussi L., Ouerfelli N., Chesneau X., Chamkha A.J., Belgacem F.B.M., Guizani A., Heat Transfer on Natural Convection in a Nanofluid Filled U-Shaped Enclosures: Numerical Investigations, *Heat Trans. Eng.*, **39**:17-29 (2018).
- [33] Snoussi L., Ouerfelli N., Sharma K.V., Vrinceanu N., Chamkha A.J., Guizani A., Numerical Simulation of Nanofluids for Improved Cooling Efficiency in a 3D Copper Microchannel Heat Sink (MCHS), *Phys. Chem. Liq.*, **57**:1–21 (2018).
- [34] Erfan-Niya H., Izadkhah M.-S., Moradkhani H., Rheological Behavior of Water–Ethylene Glycol Based Graphene Oxide Nanofluids, *Iran. J. Chem. Chem. Eng. (IJCCE)*, **37**(5):177-187 (2018).
- [35] Ali L.L., Abdel Halim S.A., Hassan Gomaa E.A., Sanad S.G., Theoretical Study of 1,4-Dioxane in Aqueous Solution and its Experimental Interaction with Nano CuSO₄, *Iran. J. Chem. Chem. Eng. (IJCCE)*, **38**(3): 43-60 (2019).
- [36] Shamsoddini R., Numerical Investigation of Fluid Mixing in a Micro-Channel Mixer with Two Rotating Stirrers by Using the Incompressible SPH Method, *Iran. J. Chem. Chem. Eng. (IJCCE)*, **36**(5): 173-183 (2017).
- [37] Ouerfelli N., Bouanz M., Excess Molar Volume and Viscosity of Isobutyric Acid + Water Mixtures Near and Far Away from the Critical Temperature, *J. Solution Chem.*, **35**:121–137 (2006).
- [38] Ouerfelli N., Iulian O., Besbes R., Barhoumi Z., Amdouni N., On the Validity of the Correlation-Belda Equation for Some Physical and Chemical Properties in 1,4-Dioxane + Water Mixtures, *Phys. Chem. Liq.*, **50**:54–68 (2012).
- [39] Das D., Ouerfelli N., The Relative Reduced Redlich-Kister and Herráez Equations for Correlating Excess properties of N,N-dimethylacetamide + formamide Binary Mixtures at Temperatures from 298.15 K to 318.15K, *J. Solution Chem.*, **41**:1334–1351 (2012).
- [40] Messaâdi A., Ouerfelli N., Das D., Hamda H., Hamzaoui A.H., Correspondence between Grunberg-Nissan, Arrhenius and Jouyban-Acree Parameters for Viscosity of Isobutyric Acid + Water Binary Mixtures from 302.15 K to 313.15 K, *J. Solution Chem.*, **41**:2186–2208 (2012).
- [41] Desnoyers J.E., Perron G., Treatment of Excess Thermodynamic Quantities for Liquid Mixtures, *J. Solution Chem.*, **26**:749–755 (1997).
- [42] Redlich O., Kister A.T., Algebraic Representation of Thermodynamic Properties and the Classification of Solutions, *Ind. Eng. Chem.*, **40**:345–348 (1948). <http://pubs.acs.org/doi/abs/10.1021/ie50458a036>

- [43] Jones G., Dole M., [The Viscosity of Aqueous Solutions of Strong Electrolytes With Special Reference to Barium Chloride](#), *J. Am. Chem. Soc.*, **51**:2950–2964 (1929).
- [44] Falkenhagen H. “[Theorie der Elektrolyte.](#)”, S. Hirzel Verlag, Leipzig (1971).
- [45] Ali A, Nain AK, Hyder S. [Ion-Solvent Interaction of Sodium Iodide and Lithium Nitrate in N,N-Dimethylformamide + Ethanol Mixtures at Various Temperatures](#), *J. Indian Chem. Soc.*, **75**(9):501–505 (1998).
- [46] Eyring H., John M.S., “[Significant Liquid Structure](#)”, John Wiley & Sons, Inc. New York, NY, USA, (1969).
- [47] Ben Haj-Kacem R., Ouerfelli N., Herráez J.V., Guettari M., Hamda H., Dallel M., [Contribution to Modeling the Viscosity Arrhenius Type-Equation for some Solvents by Statistical Correlation Analysis](#), *Fluid Phase Equilibr.*, **383**:11–20 (2014).
- [48] Ben Haj-Kacem R., Ouerfelli N., Herráez J.V., [Viscosity Arrhenius Parameters Correlation: Extension from Pure to Binary Liquid Mixtures](#), *Phys. Chem. Liq.*, **53**:776–784 (2015).
- [49] Kacem R.B., Dallel M., Al-Omair N.A., Al-Arfaj A.A., Alzamel N.O., Ouerfelli N., [Analysis of Correlation Between Viscosity Arrhenius Parameters: Extension to Ternary Liquid Mixtures](#), *Mediterr. J. Chem.*, **6**(5):208–215 (2017).
- [50] Messaâdi A., Dhouibi N., Hamda H., Belgacem F.B.M., Abdelkader Y.H., Ouerfelli N., Hamzaoui A.H., [A New Equation Correlating the Viscosity Arrhenius Temperature and the Activation Energy for Some Classical Solvents](#), *J. Chem.*, Vol. **2015**, Article ID 163262, 12 pages, (2015).
- [51] Al-Arfaj A.A., Haj-Kacem R.B., Snoussi L., Vrinceanu N., Alkhaldi M.A., Alzamel N.O., Ouerfelli N., [Correlation Analysis of the Viscosity Arrhenius-Type Equations Parameters for Some Binary Liquids Mixtures](#), *Mediterr. J. Chem.*, **6**(2):23–32 (2017).
- [52] Alzamel N.O., Alakhras F., Al-Arfaj A.A., Alkhaldi M.A., Al-Omair N.A., Al-Abbad E., Wassel A.A., Ouerfelli N., [On the Homographic Dependence of the Activation Energy and the Viscosity Arrhenius’ Temperature for Some Pure Fluids](#), *Asian J. Chem.*, **30**(09): 1937-1943 (2018).
- [53] Haj-Kacem R.B., Herráez J.V., Al-Arfaj A.A., Alkhaldi M.A., Alzamel N.O., Ouerfelli N., [Correlation Analysis of the Power Law Parameters for Viscosity of Some Engineering Fluids](#), *Phys. Chem. Liq.*, **55**:766–774 (2017).
- [54] Haj-Kacem R.B., Alzamel N.O., Al-Omair N.A., Alkhaldi M.A., Al-Arfaj A.A., Ouerfelli N., [Sensitivity of Viscosity Arrhenius-Type Equation to Density of Liquids](#), *Asian J. Chem.*, **28**(11): 2407–2410 (2016).
- [55] Kacem R.B.H., Alzamel N.O., Ouerfelli N., [Sensitivity of Viscosity Arrhenius Parameters to Polarity of Liquids](#), *Russ. J. Phys. Chem.*, **91**(9):1654–1659 (2017).